Analytical treatment of self-phase-modulation beyond the slowly varying envelope approximation

M. S. Syrchin,[1] A. M. Zheltikov,[1,2] and M. Scalora[3]

[1]*Physics Department, M.V. Lomonosov Moscow State University, 119899 Moscow, Russia*
[2]*International Laser Center, M.V. Lomonosov Moscow State University, 119899 Moscow, Russia*
[3]*Weapons Sciences Directorate, US Army Aviation and Missile Command, Huntsville, Alabama 35898-5000, USA*

Analytical treatment of the self-phase-modulation of an ultrashort light pulse is extended beyond the slowly varying envelope approximation. The resulting wave equation is modified to include corrections to self-phase-modulation due to higher-order spatial and temporal derivatives. Analytical solutions are found in the limiting regimes of high nonlinearities and very short pulses. Our results reveal features that can significantly impact both pulse shape and the evolution of the phase.

I. INTRODUCTION

With laser science breaking the femtosecond barrier, it has become more and more difficult to justify the application of the slowly varying amplitude approximation (SVEA) to explain new physical phenomena. As light pulses become increasingly shorter, new physical phenomena inaccessible to the SVEA analysis begin to play an important role, and may sometimes dominate the dynamics of ultrashort pulses and field wave forms. Recent experiments demonstrating the generation of attosecond pulses [1–3] and measurements with these pulses [4,5] raise a number of equally challenging and exciting issues related to the propagation and nonlinear-optical interactions of pulses of extremely short durations. In view of these breakthroughs, many classical results obtained in the realm of ultrafast optics may have to be revised by extending the analysis of Maxwell equations beyond the SVEA. This task becomes especially urgent when ultrashort pulses undergo nonlinear optical transformations while propagating over large distances as, for example, in conventional [6] or microstructure [7–10] fibers, or long-distance pulse propagation in the atmosphere [11,12].

Recent numerical, non-SVEA studies of propagation and nonlinear optical interactions of ultrashort light pulses have revealed new interesting scenarios of spatiotemporal dynamics and spectral evolution of ultrashort pulses [13–16], new phenomena related to the ultrafast nonlinear response of a medium [17,18], and the influence and interplay of propagation and phase-matching effects [19], to name a few. In the absence of analytical solutions it therefore seems appropriate and timely to attempt to put some of these results on a firmer theoretical ground with a systematic analytical treatment of non-SVEA effects. Accomplishing this task involves the revision of the equations of motion for basic nonlinear optical processes, such as self- and cross-phase modulation, harmonic generation, four-wave mixing, and others, with the inclusion arising from the inclusion of second-order longitudinal, spatial, and temporal derivatives.

In this paper, we provide the non-SVEA analysis specifically for self-phase-modulation (SPM) as a part of this plan, and find analytical solutions for the limiting regimes of high nonlinearities and very short pulses. Close scrutiny of these solutions then reveals important features in the dynamics of the pulse envelope and phase related to non-SVEA effects.

II. REVISED EQUATION FOR SELF-PHASE-MODULATION

We begin our analysis by writing the scalar wave equation for the electric field E in an isotropic medium with no dispersion,

$$\nabla^2 E - \frac{n^2}{c^2}\frac{\partial^2 E}{\partial t^2} = \frac{4\pi}{c^2}\frac{\partial^2 P_{nl}}{\partial t^2}, \qquad (1)$$

where n is the refractive index and c is the speed of light in the vacuum.

Without loss of generality, a short pulse propagating along the z axis, of carrier frequency ω such that $k=\omega n/c$, can be expressed as follows:

$$E(x,y,z,t) = \varepsilon(x,y,z,t)e^{i(kz-\omega t)} + \text{c.c.} \qquad (2)$$

We write the nonlinear polarization of the medium in the form of a power-series expansion, keeping the terms up to the fifth order in the field E:

$$P_{nl} = \chi^{(3)}|E|^2 E + \chi^{(5)}|E|^4 E, \qquad (3)$$

where $\chi^{(3)}$ and $\chi^{(5)}$ are the third- and fifth-order nonlinear-optical susceptibilities, respectively. In writing Eq. (2), we assume that the carrier frequency ω can be defined for our light pulse, which is true even for few-cycle pulses. In fact, Eq. (2) is quite general in that it is equivalent to a coordinate transformation. Expression (3) corresponds to the usual regime of perturbative nonlinear optics. However, our analysis will go beyond the standard SVEA treatment of self-phase-modulation. We will find non-SVEA corrections to the SPM-induced nonlinear phase shift and show that these corrections can be described in terms of the effective fifth-order nonlinear susceptibility. Since the term with the "true" fifth-order susceptibility is kept in the expression for the nonlinear polarization [Eq. (3)], we will be able to compare $\chi^{(5)}$-like non-SVEA corrections with the contribution of the true fifth-order nonlinearity.

Substituting Eqs. (2) and (3) into Eq. (1) for a plane wave, we arrive at

Report Documentation Page		Form Approved OMB No. 0704-0188

Public reporting burden for the collection of information is estimated to average 1 hour per response, including the time for reviewing instructions, searching existing data sources, gathering and maintaining the data needed, and completing and reviewing the collection of information. Send comments regarding this burden estimate or any other aspect of this collection of information, including suggestions for reducing this burden, to Washington Headquarters Services, Directorate for Information Operations and Reports, 1215 Jefferson Davis Highway, Suite 1204, Arlington VA 22202-4302. Respondents should be aware that notwithstanding any other provision of law, no person shall be subject to a penalty for failing to comply with a collection of information if it does not display a currently valid OMB control number.

1. REPORT DATE **MAY 2004**	2. REPORT TYPE **N/A**	3. DATES COVERED **-**

4. TITLE AND SUBTITLE **Analytical treatment of self-phase-modulation beyond the slowly varying envelope approximation**	5a. CONTRACT NUMBER
	5b. GRANT NUMBER
	5c. PROGRAM ELEMENT NUMBER
6. AUTHOR(S)	5d. PROJECT NUMBER
	5e. TASK NUMBER
	5f. WORK UNIT NUMBER

7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES) **Weapons Sciences Directorate, US Army Aviation and Missile Command, Huntsville, Alabama 35898-5000, USA**	8. PERFORMING ORGANIZATION REPORT NUMBER

9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES)	10. SPONSOR/MONITOR'S ACRONYM(S)
	11. SPONSOR/MONITOR'S REPORT NUMBER(S)

12. DISTRIBUTION/AVAILABILITY STATEMENT **Approved for public release, distribution unlimited**

13. SUPPLEMENTARY NOTES

14. ABSTRACT

15. SUBJECT TERMS

16. SECURITY CLASSIFICATION OF:			17. LIMITATION OF ABSTRACT **SAR**	18. NUMBER OF PAGES **6**	19a. NAME OF RESPONSIBLE PERSON
a. REPORT **unclassified**	b. ABSTRACT **unclassified**	c. THIS PAGE **unclassified**			

Standard Form 298 (Rev. 8-98)
Prescribed by ANSI Std Z39-18

$$\nabla^2 \perp \varepsilon + \frac{\partial^2 \varepsilon}{\partial z^2} + 2ik \frac{\partial \varepsilon}{\partial z} - \frac{n^2}{c^2} \frac{\partial^2 \varepsilon}{\partial t^2} + 2i\omega \frac{n^2}{c^2} \frac{\partial \varepsilon}{\partial t}$$

$$= \frac{4\pi}{c^2} \chi^{(3)} \left\{ \frac{\partial^2}{\partial t^2} - 2i\omega \frac{\partial}{\partial t} - \omega^2 \right\} |\varepsilon|^2 \varepsilon + \frac{4\pi}{c^2} \chi^{(5)}$$

$$\times \left\{ \frac{\partial^2}{\partial t^2} - 2i\omega \frac{\partial}{\partial t} - \omega^2 \right\} |\varepsilon|^4 \varepsilon. \qquad (4)$$

To represent Eq. (4) in dimensionless form, we introduce the nonlinear length $1/L_{nl} = (4\pi\omega^2 |\varepsilon_0|^2 / 2kc^2) \chi^{(3)}$, the normalized running time $\tau = (t - nz/c)/\tau_0$ (where τ_0 is the pulse duration), and the dimensionless propagation coordinate $\xi = z/L_{nl}$. We normalize the field with respect to its maximum amplitude ($\varepsilon \to \varepsilon/\varepsilon_0$). With these transformations and scalings and with an assumption that transverse effects are negligible, i.e., $\nabla^2 \perp \varepsilon = 0$, Eq. (4) can be rewritten as

$$i\frac{\partial \varepsilon}{\partial \xi} + \frac{1}{4\pi} \left\{ \frac{\lambda}{L_{nl}} \frac{\partial^2 \varepsilon}{\partial \xi^2} - \frac{\lambda}{c\tau_0} \frac{\partial^2 \varepsilon}{\partial \tau \partial \xi} \right\}$$

$$= \left\{ \left(\frac{1}{2\pi} \right)^2 \left(\frac{\lambda}{c\tau_0} \right)^2 \frac{\partial^2}{\partial \tau^2} - \frac{i}{\pi} \frac{\lambda}{c\tau_0} \frac{\partial}{\partial \tau} - 1 \right\}$$

$$\times |\varepsilon|^2 \varepsilon - \frac{|\varepsilon_0|^2 \chi^{(5)}}{\chi^{(3)}} |\varepsilon|^4 \varepsilon. \qquad (5)$$

Now we introduce the parameters $\mu_z = 1/4\pi/\lambda/L_{nl}$ and $\mu_\tau = 1/4\pi/\lambda/c\tau_0$, representing the length and time scales, respectively. Under normal circumstances, the nonlinear length is much larger than the wavelength of light. By the same token, even for a pulse only a few optical cycles in duration, μ_τ can be much smaller than unity. Therefore, keeping only terms up to the first order in μ_z and μ_τ in Eq. (5), we derive

$$i\frac{\partial \varepsilon}{\partial \xi} = -\mu_z \frac{\partial^2 \varepsilon}{\partial \xi^2} + \mu_\tau \left\{ \frac{\partial^2 \varepsilon}{\partial \xi \partial \tau} - 4i \frac{\partial (|\varepsilon|^2 \varepsilon)}{\partial \tau} \right\} - |\varepsilon|^2 \varepsilon$$

$$- \frac{|\varepsilon_0|^2 \chi^{(5)}}{\chi^{(3)}} |\varepsilon|^4 \varepsilon. \qquad (6)$$

We now focus our attention on the higher-order spatial and spatiotemporal derivatives that appear on the right-hand side of Eq. (6). Differentiating Eq. (6) with respect to z, and neglecting the terms of the order of μ_z^2, μ_τ^2, $\mu_z\mu_\tau$, and $\mu\chi^{(5)}$, we find

$$\frac{\partial^2 \varepsilon}{\partial \xi^2} = i\mu_z \frac{\partial^3 \varepsilon}{\partial \xi^3} + i2|\varepsilon| \varepsilon \frac{\partial |\varepsilon|}{\partial \xi} + i|\varepsilon|^2 \frac{\partial \varepsilon}{\partial \xi} + O(\mu_\tau)$$

$$= -|\varepsilon|^4 \varepsilon + O(\mu_\tau) + O(\mu_z). \qquad (7)$$

To estimate $\partial|\varepsilon|/\partial\xi$, we represent the field as $\varepsilon = |\varepsilon|e^{i\phi}$ and substitute this expression into Eq. (6), which yields $\partial|\varepsilon|/\partial\xi = O(\mu_\tau) + O(\mu_z)$. To find the derivative $\partial^2 \varepsilon/\partial\xi\partial\tau$, we once again differentiate Eq. (6), this time with respect to τ,

$$\frac{\partial^2 \varepsilon}{\partial \xi \partial \tau} = \frac{\partial}{\partial \tau} [i|\varepsilon|^2 \varepsilon + O(\mu_\tau) + O(\mu_z)]. \qquad (8)$$

Then, substituting Eqs. (7) and (8) into Eq. (6), and keeping the terms of the zeroth and first orders in μ_τ and μ_z, we find

$$i\frac{\partial \varepsilon}{\partial \xi} = \mu_z |\varepsilon|^4 \varepsilon - \frac{|\varepsilon_0|^2 \chi^{(5)}}{\chi^{(3)}} |\varepsilon|^4 \varepsilon - \left\{ 1 + 3i\mu_\tau \frac{\partial}{\partial \tau} \right\} |\varepsilon|^2 \varepsilon. \quad (9)$$

Expression (9) gives a SPM equation revised to include non-SVEA corrections related to higher-order derivatives in Eq. (1), as well as the term related to the fifth-order nonlinearity. With $\mu_z = \mu_\tau = 0$, Eq. (9) reduces to the standard, SVEA equation for self-phase-modulation. The revised SPM equation (9) is instructive from the physical point of view, as it provides useful insights into the physical origin of the first-order non-SVEA corrections and allows these corrections to be compared with the contribution of the fifth-order nonlinearity. The term involving μ_z gives a $\chi^{(5)}$-like, i.e., quintic in the field, correction to the SPM equation. This finding is fundamental from the viewpoint of the basic principles of nonlinear optics, as it reveals the existence of an additional physical channel whereby effects quintic in the field may contribute to the SPM of ultrashort laser pulses. In the presence of diffraction, for example, a $\chi^{(5)}$-like term would tend to defocus the beam, leading to increased nonlinear thresholds and modified spatiotemporal dynamics for long distance propagation [15]. It can be easily seen from the definition of μ_z that the role of the $\chi^{(5)}$-like term in Eq. (9) increases with the decrease in the nonlinear length L_{nl}, i.e., with the growth in the nonlinearity. Thus, $\chi^{(5)}$-like effects may influence nonlinear-optical interactions governed by the third-order nonlinearity even in the regimes of perturbative nonlinear optics. The term involving μ_τ gives a small correction to the term proportional to $|\varepsilon|^2\varepsilon$, which dominates the SPM in the SVEA regime. This correction, as it follows from the definition of the parameter μ_τ, becomes significant in the case of very short, few- and single-cycle pulses. As will be shown in the following sections, this correction may also give rise to noticeable changes in the pulse envelope and phase distribution evolution of self-phase-modulated pulses.

Expression (9) involves two terms quintic in the field E. The physical origin of these two terms is different. While the first term on the right-hand side originates from the non-SVEA correction related to the second derivative in z, the second term describes the contribution of the true fifth-order nonlinearity of the medium. It would be instructive to gain a general understanding of the relative significance of these two terms in Eq. (9) by using the relation between off-resonance nonlinear susceptibilities of different orders typical of the perturbative regime of nonlinear optics [20]: $\chi^{(n)} \sim \chi^{(1)}/|E_{at}|^{n-1}$, where E_{at} is the characteristic atomic field (typically on the order of 10^8–10^9 V/cm). The first two terms on the right-hand side of Eq. (9) can be then estimated as $|\varepsilon_0|^2 \chi^{(5)}/\chi^{(3)} \sim |\varepsilon_0|^2/|E_{at}|^2$ and $\mu_z (\pi|\varepsilon_0|^2/n^2) \chi^{(3)} \sim (\pi/n^2) \chi^{(1)} (|\varepsilon_0|^2/|E_{at}|^2) \sim |\varepsilon_0|^2/|E_{at}|^2$. The non-SVEA correction in a nonresonant situation is thus of the same order of smallness as the term related to the true fifth-order nonlinearity. These two channels of nonlinear-optical interactions can, therefore, interfere either constructively or destructively, giving rise to interesting effects, enhancing or suppressing nonlinear phenomena, depending on the relative sign of the first two terms in Eq. (9). In what follows, we will present an iterative procedure, allowing an analytical integration of non-SVEA SPM equations for the amplitude and the phase of ultrashort pulses including only the cubic optical nonlinearity.

III. ANALYTICAL SOLUTIONS TO THE NON-SVEA SPM EQUATION

In this section, we will develop, following the plan outlined above, an iterative procedure solving the generic non-SVEA SPM equation (9) for a medium with cubic and quintic nonlinearities. Representing the field of a light pulse as $\varepsilon(\xi, \tau) = A(\xi, \tau) \exp[i\phi(\xi, \tau)]$, we arrive at the following set of equations for the amplitude A and the phase ϕ of the field:

$$\frac{\partial A}{\partial \xi} = -9\mu_\tau A^2 \frac{\partial A}{\partial \tau},$$

$$\frac{\partial \phi}{\partial \xi} = A^2 - \bar{\mu}_z A^4 - 3\mu_\tau A^2 \frac{\partial \phi}{\partial \tau},$$

where $\bar{\mu}_z = \mu_z - |\varepsilon_0|^2 \chi^{(5)}/\chi^{(3)}$.

We now write the field amplitude as a series expansion in the small parameter μ_τ,

$$A = A_{(0)} + \mu_\tau A_{(1)} + \mu_\tau^2 A_{(2)} + \cdots, \tag{11}$$

where $A_{(0)}$ is the SVEA solution to the SPM equation for the field amplitude,

$$\frac{\partial A_{(0)}}{\partial \xi} = 0, \tag{12}$$

and $A_{(1)}$ and $A_{(2)}$ are the non-SVEA corrections of the first and second orders in μ_τ, respectively. The equation for the first-order non-SVEA correction to the field amplitude is then written as

$$\frac{\partial A_{(1)}}{\partial \xi} = -9A_{(0)}^2 \frac{\partial A_{(0)}}{\partial \tau}. \tag{13}$$

The solution to Eq. (12) is well known [20]: $A_{(0)}(\xi, \tau) = A_0(\tau)$, where $A_0(\tau)$ is the initial pulse envelope normalized to the maximum field amplitude $[A_0(\tau) = \exp(-\tau^2)$ for a Gaussian pulse]. Integration of Eq. (13) yields

$$A_{(1)}(\xi, \tau) = -9A_0^2 \frac{\partial A_0}{\partial \tau} \xi. \tag{14}$$

The solution to the non-SVEA SPM equation (9) in the first-order approximation in μ_τ is thus written as

$$A(\xi, \tau) = A_0 - 9\mu_\tau A_0^2 \frac{\partial A_0}{\partial \tau} \xi, \quad A_0 \equiv A_0(0, \tau). \tag{15}$$

We can now proceed with the solution of the equation for the phase in the set of Eqs. (10). We represent the phase as

$$\phi = \phi_{(0)} + \mu_\tau \phi_{(1)} + \mu_\tau^2 \phi_{(2)} + \cdots \tag{16}$$

and take the solution to the equation

$$\frac{\partial \phi_{(0)}}{\partial \xi} = A^2 - \bar{\mu}_z A^4 \tag{17}$$

as the zeroth-order iteration. In view of Eq. (17), the small parameter $\bar{\mu}_z$ also appears in the series expansion (16) for the phase. Substituting Eq. (16) into the second equation in the set (10), we derive the following equation for the first-order non-SVEA correction to the phase:

$$\frac{\partial \phi_{(1)}}{\partial \xi} = -3A^2 \frac{\partial \phi_{(0)}}{\partial \tau}. \tag{18}$$

Substituting the first-order non-SVEA solution (15) for the amplitude into Eq. (18), keeping only the terms of the zeroth and first orders in μ_τ and $\bar{\mu}_z$ in the resulting equation, and assuming that the input pulse has no initial chirp, we arrive at the following solution for the phase:

$$\phi_{(0)}(\xi, \tau) = \left(A_0^2 - \bar{\mu}_z A_0^4 - 18\mu_\tau A_0^3 \frac{\partial A_0(\tau)}{\partial \tau} \xi \right) \xi,$$

$$\phi_{(1)}(\xi, \tau) = -6A_0^3 \frac{\partial A_0(\tau)}{\partial \tau} \xi^2.$$

Combining the solutions for the phase and the amplitude given by Eqs. (15) and (19), we derive the following expression for the field:

$$\varepsilon(\xi, \tau) = A_0 \left[1 - 9\mu_\tau A_0 \frac{\partial A_0}{\partial \tau} \xi \right] \exp[i(\phi_{\text{SVEA}} - \Delta\phi_z - \Delta\phi_\tau)], \tag{20}$$

where

$$\phi_{\text{SVEA}} = A_0^2 \xi, \tag{21}$$

is the SVEA solution for the SPM-induced phase shift,

$$\Delta\phi_\tau = 24\mu_\tau A_0^3 \frac{\partial A_0(\tau)}{\partial \tau} \xi^2 \tag{22}$$

is the non-SVEA correction to the nonlinear phase shift related to the second derivative in time, and

$$\Delta\phi_z = \bar{\mu}_z A_0^4 \xi \tag{23}$$

is the non-SVEA correction originating from the second derivative in z.

For a pulse with an initial phase modulation $\phi(0, \tau) \equiv \phi_0(\tau) = \alpha\tau^2$ $(\alpha\mu_\tau \ll 1)$, the correction to the nonlinear phase shift is given by

$$\Delta\phi_\tau = 24\mu_\tau A_0^3 \frac{\partial A_0(\tau)}{\partial \tau} \xi^2 - 6\mu_\tau A_0^2 \alpha. \tag{24}$$

Expression (23) suggests that the non-SVEA correction $\Delta\phi_z$ to the SPM-induced phase shift is quartic in the field. This functional dependence is typical of $\chi^{(5)}$ nonlinear-optical effects. In our case, however, the smallness of the phase shift $\Delta\phi_z$ is related not only to the fifth-order susceptibility $\chi^{(5)}$, but is governed by the parameter $\bar{\mu}_z$, which is in turn controlled by both the third- and fifth-order susceptibilities $\chi^{(3)}$ and $\chi^{(5)}$. In a practically important situation of a medium with a resonance-enhanced third-order susceptibility $\chi^{(3)}$, the phase shift $\Delta\phi_z$, given by Eq. (23) can, therefore, noticeably exceed the true $\chi^{(5)}$-induced phase shifts, giving rise, as will be shown below, to detectable (and sometimes
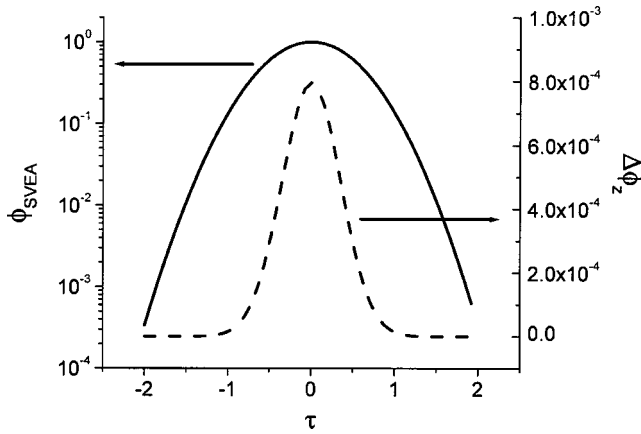
FIG. 1. The nonlinear phase shift ϕ_{SVEA} calculated within the framework of the slowly varying envelope approximation (solid line) and the non-SVEA correction $\Delta\phi_z$ to the nonlinear phase shift related to the second-order derivative in z (dashed line) for $\xi=1$ with $\bar{\mu}_z=10^{-3}$. The variable τ is normalized to the pulse duration.
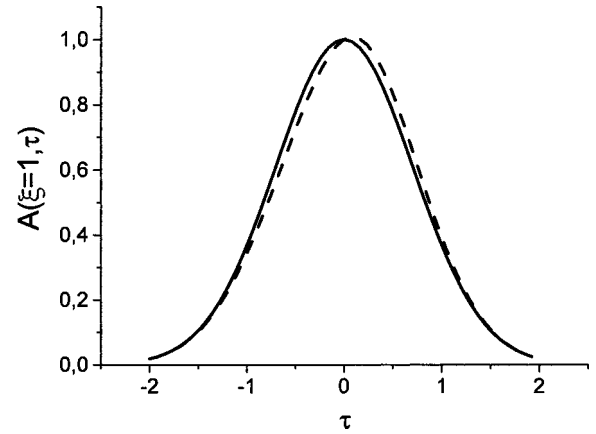

FIG. 2. The envelope of a self-phase-modulated pulse calculated within the framework of the slowly varying envelope approximation (solid line) and with the use of Eq. (20) (dashed line), including effects related to the second-order derivative in τ, for $\xi=1$ with $\mu_\tau=10^{-2}$, $\bar{\mu}_z\gg\mu_\tau$. The variable τ is normalized to the pulse duration.

significant) $\chi^{(5)}$-like corrections to the phase in the purely perturbative regime of nonlinear-optical interactions.

Non-SVEA effects related to the second-order time derivative, as can be seen from Eqs. (22) and (24), give rise to distortions in the envelope of a light pulse, which becomes z dependent even in the absence of group-velocity dispersion. Expressions (22)–(24) are also instructive in demonstrating that both pulse-envelope and phase-profile distortions related to the non-SVEA effects accumulate with z as the pulse propagates through the nonlinear medium. This dependence on the longitudinal coordinate can lead to noticeable deviations from the SVEA results for pulse envelopes and nonlinear phase shifts for sufficiently large propagation distances, as, for example, in the case of standard [6] or microstructure optical fibers [7–10] or nonlinear-optical interactions in the atmosphere [11,12].

IV. RESULTS AND DISCUSSION

To assess the role of non-SVEA effects in self-phase-modulation, we use the solutions to the revised SPM equation derived above to calculate the non-SVEA phase shifts and distortions in pulse envelopes for different values of parameters $\bar{\mu}_z$ and μ_τ. Figure 1 displays the nonlinear phase shift ϕ_{SVEA} calculated within the framework of the standard SVEA approach (solid line) and the non-SVEA correction $\Delta\phi_z$ to the nonlinear phase shift related to the second-order derivative in z (dashed line) for $\xi=1$. The non-SVEA phase shift was calculated using Eq. (23) with $\bar{\mu}_z\sim10^{-3}$ (which corresponds to $L_{nl}=100$ μm in a medium with $\chi^{(5)}=0$). Such values of $\bar{\mu}_z$ can easily be achieved, for example, by propagating light pulses with a wavelength of 1 μm and the intensity on the order of 10^{12} W/cm^2 through fused silica (in the absence of true fifth-order nonlinearity). The non-SVEA correction to the phase shift, $\Delta\phi_z$, as can be seen from Fig. 1, can reach a maximum of about 0.1 % under these conditions. This part of the phase shift, however, increases proportionally to $\bar{\mu}_z$, reaching 0.1 % for $\bar{\mu}_z=0.01$. These estimates

show that non-SVEA phase shifts may play an important role, for example, in nonlinear-optical experiments in microstructure fibers [10], where light intensities exceeding 10^{13} W/cm^2 are often achieved by coupling femtosecond pulses in a few-micron fiber core.

In the regime of very short pulses, with $\bar{\mu}_z\gg\mu_\tau$, non-SVEA pulse-envelope and phase-profile distortions accumulate as the pulse propagates through a medium. Figure 2 shows distortions in the envelope of a light pulse with an initial pulse duration of about 30 fs caused by effects related to the second-order time derivative in the wave equation. The changes in the pulse envelope are already quite noticeable for $\xi=1$. The influence of non-SVEA phase shifts is illustrated in Fig. 3, which compares the nonlinear phase shift of a pulse with a duration of about 30 fs calculated within the framework of the SVEA approach with the nonlinear phase shift calculated with the use of Eqs. (20) and (22), including
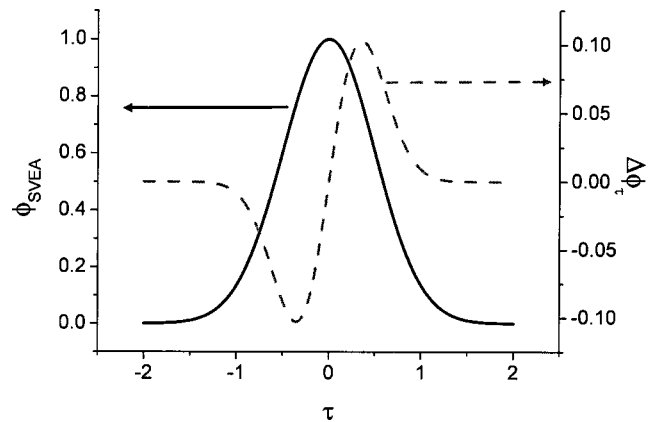

FIG. 3. The nonlinear phase shift ϕ_{SVEA} calculated within the framework of the slowly varying envelope approximation (solid line) and the correction to the phase shift $\Delta\phi_\tau$ calculated with the use of Eq. (22) (dashed line), including effects related to the second-order derivative in τ, for $\xi=1$ with $\mu_\tau=10^{-2}$. The variable τ is normalized to the pulse duration.
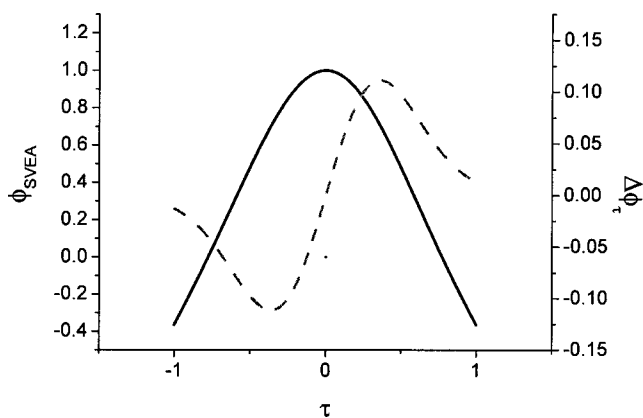
FIG. 4. The nonlinear phase shift of a short pulse with an initial linear chirp calculated within the framework of the slowly varying envelope approximation (solid line) and the correction to the phase shift $\Delta\phi_\tau$ calculated with the use of Eq. (24) (dashed line), including effects related to the second-order derivative in τ, for $\xi=1$ with $\alpha=-0.5$ and $\mu_\tau=10^{-2}$.

non-SVEA corrections related to the second-order time derivative. Similar to pulse-envelope distortions, the deviation of the non-SVEA nonlinear phase shift from the SVEA result becomes quite noticeable already for $\xi=1$, increasing in the process of pulse propagation in accordance with Eqs. (20) and (22).

Putting an initial chirp on the pulse adds phase-control aspects to the analysis of nonlinear interactions of ultrashort pulses. Figures 4 and 5 show that such an analysis should include non-SVEA effects, which may have a noticeable influence on phase-controlled nonlinear-processes. Figure 4 compares the SVEA nonlinear phase shift of a short pulse having an initial linear chirp with a chirp parameter $\alpha= -0.5$ and $\mu_\tau=10^{-2}$ with the results of calculations performed with the use of Eq. (24), including effects related to the second-order time derivative. The evolution of the nonlinear phase shifts for transform-limited and initially chirped pulses illustrated in Figs. 5(a)–5(c) shows how the non-SVEA part of the nonlinear shift is accumulated and the distribution of the nonlinear shift within the pulse becomes more and more asymmetric as the pulse propagates through the nonlinear medium. Non-SVEA phase corrections should be, therefore, taken into consideration in coherent-control experiments [21–26], where chirped ultrashort laser pulses are used to steer molecular excitations, wave-packet motions, chemical reactions, or nonlinear-optical interactions. Non-SVEA effects, as can be seen from Fig. 5, also noticeably influence the SPM-phase-shift precompensation function of the initial chirp, as the result of such a precompensation may deviate from expectations based on the SVEA analysis. Positive initial chirp, as can be seen from the comparison of the evolutions of the corrections $\Delta\phi_\tau$ for transform-limited and initially chirped pulses shown by the dashed and dotted lines in Figs. 5(a)–5(c), respectively, allows phase-profile distortions related to non-SVEA effects to be reduced or precompensated. This precompensation is effective, however, only within propagation lengths on the order of the nonlinear
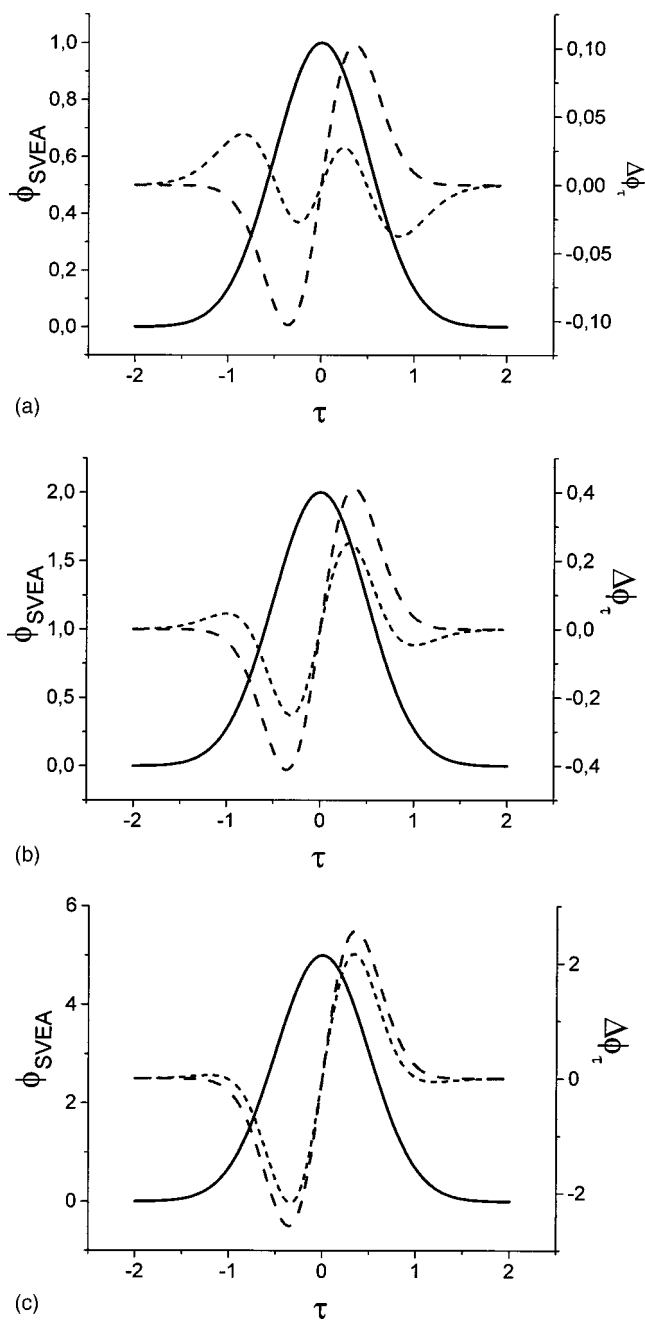

(a)

(b)

(c)

FIG. 5. The nonlinear phase shift for an initially transform-limited pulse calculated within the framework of the slowly varying envelope approximation (solid lines) and the correction to the phase shift $\Delta\phi_\tau$ calculated with the use of Eq. (24) for an initially transform-limited pulse with $\alpha=0$ (dashed lines) and a pulse having a linear initial chirp with $\alpha=0.5$ (dotted lines) for $\xi=1$ (a), $\xi=2$ (b), and $\xi=5$ (c); $\mu_\tau=10^{-2}$.

length [Fig. 5(a)]. Non-SVEA phase-profile distortions accumulate as the pulse propagates through the medium [Figs. 5(b) and 5(c)], the first term in Eq. (24) starts to dominate, and the expression for the non-SVEA phase-shift correction $\Delta\phi_\tau$ is reduced to Eq. (22). This analysis shows that a properly chosen initial chirp of short pulses may serve as an important knob in phase-controlled molecular motions and nonlinear interactions.

V. CONCLUSION

Thus, analysis performed in this paper shows that the scenarios of nonlinear-optical interactions of ultrashort pulses may sometimes noticeably differ from predictions of the slowly varying envelope approximation. We have derived a revised equation for self-phase modulation, including corrections related to higher-order derivatives in the wave equation. We have also developed an iterative procedure allowing the analytical solution of this equation for the limiting regimes of high nonlinearities and very short pulses. Analysis of these solutions reveals interesting features in the pulse-envelope and phase-profile evolution related to non-SVEA effects in self-phase-modulation. It was shown, in particular, that $\chi^{(5)}$-like effects, i.e., effects of the fifth order in the laser field, may influence nonlinear-optical interactions governed by the third-order nonlinearity even in the regimes of perturbative nonlinear optics. Many of the non-SVEA effects accumulate with the propagation coordinate, leading to noticeable changes in the pulse envelope and the phase distribution in the laser pulse for large propagation lengths, e.g., in optical fibers or in nonlinear-optical interactions in the atmosphere. Non-SVEA phase corrections and pulse-envelope distortions should be taken into consideration in coherent-control experiments, where chirped ultrashort laser pulses are used to steer molecular excitations, wave-packet motions, chemical reactions, or nonlinear-optical interactions.

[1] P. M. Paul, E. S. Toma, P. Breger, G. Mullot, F. Augé, Ph. Balcou, H. G. Muller, and P. Agostini, Science **292**, 1689 (2001).

[2] M. Drescher, M. Hentschel, R. Kienberger, G. Tempea, Ch. Spielmann, G. A. Reider, P. B. Corkum, and F. Krausz, Science **291**, 1923 (2001).

[3] M. Hentschel, R. Kienberger, Ch. Spielmann, G. A. Reider, N. Milosevic, T. Brabec, P. Corkum, U. Heinzmann, M. Drescher, and F. Krausz, Nature (London) **414**, 511 (2001).

[4] M. Drescher, M. Hentschel, R. Kienberger, M. Uiberacker, V. Yakovlev, A. Scrinzi, Th. Westerwalbesloh, U. Kleineberg, U. Heinzmann, and F. Krausz, Nature (London) **419**, 803 (2002).

[5] R. Kienberger, M. Hentschel, M. Uiberacker, Ch. Spielmann, M. Kitzler, A. Scrinzi, M. Wieland, Th. Westerwalbesloh, U. Kleineberg, U. Heinzmann, M. Drescher, and F. Krausz, Science **297**, 1144 (2002).

[6] G. P. Agrawal, *Nonlinear Fiber Optics* (Academic, San Diego, 2001).

[7] J. C. Knight, T. A. Birks, P. St.J. Russell, and D. M. Atkin, Opt. Lett. **21**, 1547 (1996); J. C. Knight, J. Broeng, T. A. Birks, and P. St.J. Russell, Science **282**, 1476 (1998).

[8] P. J. Bennet, T. M. Monro, and D. J. Richardson, Opt. Lett. **24**, 1203 (1999); T. M. Monro, P. J. Bennett, N. G.R. Broderick, and D. J. Richardson, *ibid.* **25**, 206 (2000).

[9] A. B. Fedotov, A. M. Zheltikov, L. A. Mel'nikov, A. P. Tarasevitch, and D. von der Linde, JETP Lett. **71**, 281 (2000); M. V. Alfimov, A. M. Zheltikov, A. A. Ivanov, V. I. Beloglazov, B. A. Kirillov, S. A. Magnitskii, A. V. Tarasishin, A. B. Fedotov, L. A. Mel'nikov, and N. B. Skibina, *ibid.* **71**, 489 (2000).

[10] *Nonlinear Optics of Photonic Crystals*, feature issue of J. Opt. Soc. Am. B **19** (9) (2002), edited by C. M. Bowden and A. M. Zheltikov.

[11] B. La Fontaine, F. Vidal, Z. Jiang, C. Y. Chien, D. Comtois, A. Desparois, T. W. Johnson, J.-C. Kieffer, and H. Pépin, Phys. Plasmas **6**, 1615 (1999).

[12] J. Kasparian, R. Sauerbrey, D. Mondelain, S. Niedermeier, J. Yu, J.-P. Wolf, Y.-B. André, M. Franco, B. Prade, S. Tzortzakis, A. Mysyrowicz, M. Rodriguez, H. Wille, and L. Wöste, Opt. Lett. **25**, 1397 (2000).

[13] T. Brabec and F. Krausz, Rev. Mod. Phys. **72**, 545 (2000).

[14] A. L. Gaeta, Phys. Rev. Lett. **84**, 3582 (2000).

[15] N. Akozbek, M. Scalora, C. M. Bowden, and S. L. Chin, Opt. Commun. **191**, 353 (2001).

[16] S. Coen, A. Hing Lun Chau, R. Leonhardt, J. D. Harvey, J. C. Knight, W. J. Wadsworth, and Ph. St.J. Russell, J. Opt. Soc. Am. B **19**, 753 (2002).

[17] R. W. Ziolkowski, J. M. Arnold, and D. M. Gogny, Phys. Rev. A **52**, 3082 (1995).

[18] A. V. Tarasishin, S. A. Magnitskii, V. A. Shuvaev, and A. M. Zheltikov, Opt. Express **8**, 452 (2001).

[19] N. Akozbek, A. Iwasaki, A. Becker, M. Scalora, S. L. Chin, and C. M. Bowden, Phys. Rev. Lett. **89**, 143901 (2002).

[20] Y. R. Shen, *The Principles of Nonlinear Optics* (Wiley, New York, 1984).

[21] B. Kohler, V. V. Yakovlev, J. Che, J. L. Krause, M. Messina, K. R. Wilson, N. Schwentner, R. M. Whitnell, and Y. Yan, Phys. Rev. Lett. **74**, 3360 (1995).

[22] A. Assion, T. Baumert, J. Helbing, V. Seyfried, and G. Gerber, Chem. Phys. Lett. **259**, 488 (1996).

[23] G. Cerullo, C. J. Bardeen, Q. Wang, and C. V. Shank, Chem. Phys. Lett. **262**, 362 (1996).

[24] V. V. Yakovlev, C. J. Bardeen, J. Che, J. Cao, and K. R. Wilson, J. Chem. Phys. **108**, 2309 (1998).

[25] D. Zeidler, S. Frey, K.-L. Kompa, and M. Motzkus, Phys. Rev. A **64**, 023420 (2001).

[26] J. L. Herek, W. Wohlleben, R. J. Cogdell, D. Zeidler, and M. Motzkus, Nature (London) **417**, 533 (2002).